Polonium 210 in Tobacco, Cigarette Smoke, and Selected Human Organs

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THE POLONIUM 210 in tobacco may be implicated in the origin of lung cancer (1,2). Speculation on this possibility prompted a study at the Northeastern Radiological Health Laboratory, Public Health Service, to determine the levels of polonium 210 in several brands of cigarettes. The levels of lead 210 and radium 226 were also measured to determine whether the polonium 210 was in equilibrium with these precursors or was present independently. Samples of human organs were also analyzed for polonium 210 content to determine whether a difference could be detected in the concentration of this radionuclide between smokers and nonsmokers.

Radionuclides in Tobacco

The tobacco products chosen for the study included nonfiltered and filtered cigarettes and cigarettes treated for the removal of tars and nicotine. Although not now normally considered as great a health hazard to the lungs because of the method of smoking, cigars and pipe tobacco were also assayed for polonium 210 content.

Methodology of analysis. The following methodology was used for determining radium 226, lead 210, bismuth 210, and polonium 210 in tobacco:

Radium 226 was determined by dissolving the tobacco ash in hydrochloric acid, and the nuclide was determined by emanation, collection, and counting of the gaseous daughter radon 222 (3).

Mrs. Ferri is a chemist, and Mr. Baratta is chief of Analytical Services, Northeastern Radiological Health Laboratory, Division of Radiological Health, Public Health Service, Winchester, Mass. To determine lead 210, the tobacco ash was dissolved in hydrochloric acid with carrier present, passed over an anion exchange column to remove bismuth, and precipitated as the sulfide. The sulfide was dissolved, collected on an anion exchange column, eluted, precipitated as the chromate, and counted. The ingrowth of bismuth 210, daughter of lead 210, was observed for calculations (as outlined in personal communication from Paul B. Hahn, chemist, Northeastern Radiological Health Laboratory).

To determine bismuth 210, the tobacco ash was dissolved in hydrochloric acid, carrier added, and the bismuth collected on an anion column. It was eluted, precipitated as the oxychloride, and counted. The decay of the 5-day bismuth 210 was observed for purity of recovery (personal communication from Hahn).

Because polonium 210 is volatile at dry ashing temperatures, the samples for analysis were wetashed with nitric acid and perchloric acid; the resulting solution was made 0.5N in hydrochloric acid. The polonium was spontaneously deposited on a silver disk, counted for alpha activity in a gasflow proportional counter, and checked for purity on an alpha spectrometer (4).

Results of analysis. Our analyses clearly demonstrated that daughter products lead 210, bismuth 210, and polonium 210 in tobacco are not in equilibrium with the parent, radium 226 (table 1). The high lead-to-radium ratios indicate that lead 210 reaches the tobacco by direct uptake from the soil and air rather than merely as the daughter of radium 226 in the tobacco.

Marsden (5) conducted experiments with autoradiographic techniques to measure the ac-

tivity of upper and lower surfaces of tobacco leaves, and stated that the effect of polonium fallout is minor compared with activity derived from the soil through roots. Gregory (6) stated the same uncertainty as to the derivation of lead 210 in tobacco but suggested an insular or coastal region for growing might greatly disburse the radon, which emanates from the soil, and result in less fallout of decay products for that area.

The values obtained for radium 226 and polonium 210 agree with those reported for various domestic tobaccos by Tso and co-workers (7). Lead 210 and polonium 210 were not in equilib-

Table 1. Radionuclides in tobacco, picocuries per cigarette (±2 standard deviations), per cigar, and per gram of pipe tobacco

Type	¹ Radium 226	¹ Lead 210	¹ Bismuth 210	¹Polonium 210	Ratio of lead to radium
Cigarettes: Nonfiltered, brand A	$\begin{array}{c} 0.\ 15\ \pm.\ 01(1)\\ .15\ \pm.\ 01(1)\\ .17\ \pm.\ 01(3)\\ .12\ \pm.\ 01(1)\\ .11\ \pm.\ 01(2)\\ .11\ \pm.\ 01(2)\\ .74\ \pm.\ 10(2)\\ .096\pm.\ 01(1) \end{array}$	$\begin{array}{c} .42 \pm .04(3) \\ .33 \pm .04(1) \\ ^{2}.49 \\ ^{2}.33 \\ ^{2}.35 \\ ^{2}3.55 \end{array}$	$.40 \pm .04(4)$	$egin{array}{c} .\ 32 \pm .\ 04(13) \ .\ 48 \pm .\ 04(2) \ .\ 34 \pm .\ 04(3) \ .\ 34 \pm .\ 04(2) \ 3.\ 60 \pm .\ 05(2) \ \end{array}$	2. 9 2. 8 1. 9 4. 1 3. 0 3. 2 4. 0 4. 3

¹ Number of samples in parentheses.

² Assumed from bismuth 210 concentration.

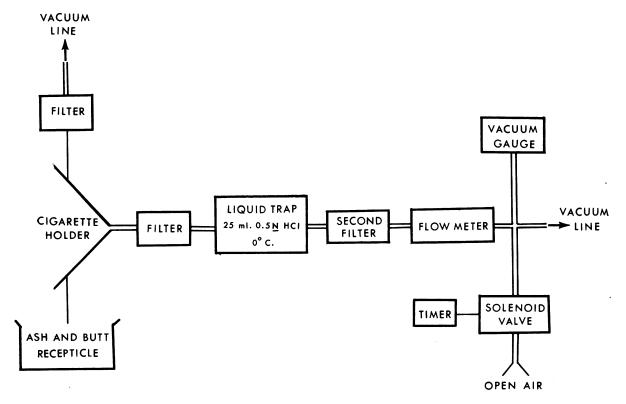


Figure 1. Flow diagram of smoking machine at Northeastern Radiological Health Laboratory,
Public Health Service

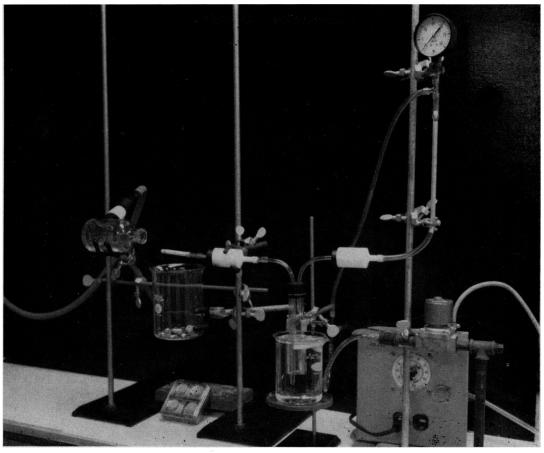


Figure 2. Smoking machine at Northeastern Radiological Health Laboratory,
Public Health Service

rium in the pipe tobacco. This may indicate that the tobacco preferentially takes up lead 210 and perhaps reaches the market before equilibrium can be established. On the other hand, the subsequent "aging" time for cigarette tobacco of approximately 2 years allows lead 210 and its daughters bismuth 210 and polonium 210 to reach equilibrium.

Use of Smoking Machine

Nuclides that are volatile at the temperature of burning tobacco are of primary interest as a potential health hazard. Before this hazard can be assessed, it is necessary to determine the amount of radionuclides inhaled in the smoke and deposited in the mouth, bronchial tract, and lungs of the smoker.

To collect the smoke that is normally inhaled, a "smoking machine" was designed and tested at our laboratory (figs. 1, 2). The polonium 210 that has volatilized and the smoke and tars are collected on two filters and a hydrochloric acid trap. The airflow is regulated by a solenoid valve that intermittently connects the system to a vacuum line to simulate puffing. The four primary operating characteristics of the smoking machine are:

- 10 puffs per cigarette.
- 3 seconds per puff.
- 26-second intervals between puffs.
- Flow rate: 18 milliliters per second, standard temperature and pressure.

Sidestream smoke is collected by placing a bottle over the burning cigarette. A filter, connected to a vacuum line, is attached to the bottle. Thus when smoke is not being drawn through the cigarette, it collects on the filter in the vacuum flask.

The smoking machine was compared with the actual smoking of three persons. The polonium

210 in butts and ashes from one package (20 cigarettes) each of the nonfilter, filter, and treated-tobacco types smoked by people were compared with the butts and ashes of cigarettes processed in the smoking machine (table 2). Our comparison indicated that the smoking machine is a good approximation of actual smoking and can be used to determine the distribution of radionuclides between cigarette smoke and residue.

Polonium 210 in Cigarette Smoke

The technique used for determining polonium 210 in the various fractions of the smoked cigarette includes plating the radionuclide onto silver disks. To evaluate the efficiency of plating, the procedure was studied, and the following plating efficiency readings were obtained:

Plating	g efficiency
Sample (pe	ercent)
Standard	99. 5
Total cigarettes	94. 5
Traps	100.0
Butts and ashes	93. 5
Average ±2 standard deviations	96.9 ± 3.6

Before plating, 200 counts per minute of polonium 210 were added to each cigarette sample previously depleted of the polonium. The recoveries from the various media demonstrated that there was no significant loss of polonium 210 during the plating process.

Table 2. Percent of polonium 210 in cigarette butts, ashes, and filters from smokers and the smoking machine

Type of cigarette	Smokers	Smoking machine	
NonfilteredFiltered, including filter Treated, including filter	48 50 51	47 53 50	

With the smoking machine, material balances for polonium 210 were obtained on four major brands of cigarettes: filtered, nonfiltered, filtered treated tobacco, and a filtered brand containing pipe tobacco. The unsmoked cigarette was analyzed for polonium 210 content. The smoked cigarette was analyzed for polonium 210 content in the (a) smoke inhaled, (b) sidestream smoke (smoke not inhaled), (c) unsmoked tobacco and ash, and (d) filter.

Each analysis was performed on the smoke and residue from 20 cigarettes. Several other brands of cigarettes were analyzed for polonium 210 content in the total cigarette and in the inhaled smoke. The reported values represent the maximum total activity inhaled in the smoke (table 3). To normalize the data for varying quantities of tobacco in each cigarette, the results are given in picocuries per cigarette and picocuries per gram of tobacco. No attempt was

Table 3. Polonium 210 material balance in 1 cigarette and in 1 gram of tobacco, in picocuries 1

	Brand B 2		Brand C 3		Brand D 4		Brand E 5		Brand F 6	
Source of sample		Gram of tobacco		Gram of tobacco		Gram of tobacco		Gram of tobacco		Gram of tobacco
Total cigarette Inhaled smoke	0. 370 . 082	0. 411 . 091	0. 320 . 049	0. 403 . 061	0. 480 . 087	0. 560 . 102	0. 340 . 037	0. 410 . 045	0. 342 . 105 (30. 7)	0. 357 . 109
Sidestream smoke	(22.2) $.091$ (24.5)	. 100	(15.1) 105 (32.9)	. 133	(18. 1)		(11. 0) . 139 (40. 8)	. 168	. 111 (32. 4)	. 116
Butt and ash	. 172	. 191	. 124 (38. 8)	. 157			. 171 (50. 3)	. 206	.110 $(32, 2)$. 115
Filter			.044 (13.7)	. 055					.030 (8.8)	. 031
Material balance, percent	93. 4		100. 5				102. 1		104. 1	

¹ Percent activity in parentheses.

² Nonfiltered.

³ Filtered, cellulose.

⁴ Filtered, cellulose and charcoal.

⁵ Filtered, cellulose and charcoal treated.

⁶ Filtered, cellulose, pipe tobacco.

made to determine the polonium 210 content in the exhaled smoke or reaching the mouth or lungs via the inhaled smoke.

Comparison of polonium activity in smoke and residues of filtered and nonfiltered cigarettes showed that variations in the portions inhaled have no bearing on the total activity in the cigarette. For example, brand F, containing 29.8 percent less activity in the total cigarette than brand D, showed more activity in the inhaled-smoke portion. Similarly, the inhaled smoke from brands D and B contained about the same level of activity, with about a 23 percent difference in available activity in the total cigarette. It would therefore appear that different filtering mechanisms, whether of artificial material or real tobacco, and the cut are contributing to the variation in activities found in inhaled smoke. Although most cigarettes contain a mixture of tobaccos, an average of 0.43 picocuries per gram of tobacco was obtained for all brands, which compares well with the average value of 0.49 picocuries per gram reported by Gregory (6) for "burley" grown in the United States.

Radford and Hunt (1) reported an average value of 0.425 picocuries per total cigarette and an 85 percent material balance on smoke and residues. We observed an average value 15 percent lower (0.37 picocuries per total cigarette) for all brands tested and material balances of approximately 100 percent (table 3). If all analyses of inhaled smoke from filtered cigarettes were averaged and normalized as picocuries per gram of tobacco, a value of 0.079 would be obtained, compared with 0.091 for nonfiltered cigarettes—a 15 percent difference. In the same

manner, if the residues (butt, ash, and filter where applicable) of filtered and nonfiltered cigarettes were compared, about a 20 percent difference would be observed.

The greatest variable between types of cigarettes appears to be in the sidestream smoke, which in filtered cigarettes is 40 pecent higher. Thus the question is raised whether the filter causes the tobacco to burn at a greater rate when it is not being puffed. In this laboratory we observed, with human smokers, that a backstream of smoke was visible from the mouth end of the cigarette after the smoker had finished a puff. The stream appeared to be much heavier and of longer duration from brands B, D, and F than from brands C and E when all the brands were smoked by several persons. If this also happens when using a smoking machine, some residual backstream smoke is entering as mainstream smoke and could not be considered as sidestream smoke. This could account for the variations of higher activity in the sidestream smoke and less activity in the mainstream smoke from certain brands.

Average Calculated Dose to the Lung

Tobacco smoke has been found to contain particles ranging from 0.01μ to 0.25μ in diameter (8). By using this information and assuming that particles less than 0.5μ in diameter have ready access to the alveoli so as to provide a radiation dose distributed uniformly throughout the lung lobes (9), doses for the whole lung as a critical organ were calculated (10). Calculations were based on numbers obtained from the smoking machine. We assumed that each day a person regularly smoked two packages

Table 4. Dose of polonium 210 to the lung by smoking two packs of cigarettes daily

Type of cigarette	Polonium 210, picocuries inhaled per cigarette	Dose to lungs, millirems per year	Actual intake, microcuries per week	Actual intake of MPC ¹ (percent)	
Brand B (nonfiltered)	0. 082 . 049 . 037 . 105	73 41 33 92	$\begin{array}{c} 2.39 \times 10^{-5} \\ 1.37 \times 10^{-5} \\ 1.04 \times 10^{-5} \\ 2.94 \times 10^{-5} \end{array}$	3. 4 1. 9 1. 5 4. 2	

¹ Maximum permissible concentration of polonium 210 in air per person in the general population, with lung as the critical organ=7.06×10⁻⁴ microcuries per week. Source: National Bureau of Standards Handbook No. 69, August 1963.

of filtered or nonfiltered cigarettes. We also assumed an average of 1,200 grams for the total mass of the right and left lung, an effective half life of 18 days (11), a 100 percent fraction inhaled and reaching the lung, and that polonium 210 equilibrium had previously been reached in approximately 78 days of prior smoking.

Based on these assumptions, the calculated dose was 41 millirems per year from the cellulose filtered brand; 73 millirems per year from the nonfiltered brand; 33 millirems per year from the filtered (cellulose and charcoal) treated-tobacco cigarettes; and 95 millirems per year from the filtered cigarette containing pipe tobacco (table 4). These calculated intakes are 1.5 to 4.2 percent of the maximum permissible concentration for polonium 210 in air per person in the general population, with the lung as the organ of reference (10a).

Hatch and Gross (8a) have stated that 75 percent of the inhaled material is deposited in respiratory uptake, 50 percent in the nasal chamber and upper respiratory tract, and 25 percent in the lungs. Half of the amount in the lungs is cleared from the lungs in 24 hours, and the remaining 12.5 percent is transferred from the lungs to the bloodstream. Therefore, these dosage figures could be high by as much as a factor of 8. It is important to note the above calculations refer to the average dose over the entire lung tissue. Other authors (1) have pointed out that nonhomogeneous deposition may result in localized doses much greater than reported in this paper.

Polonium 210 in Human Organs

To determine if polonium 210 from smoking concentrates in various organs of the human body, specimens of lungs and other soft tissue of a smoker and a nonsmoker were analyzed and compared (table 5). Soft tissue chosen for the study were liver, kidney, heart, and psoas muscle. Lungs were dissected into parenchyma, nodes, and bronchi. The age of the smokers ranged from 36 to 75 years; of nonsmokers, 52 to 73 years. Smokers as referred to in table 5 were composed of cigar, pipe, and cigarette smokers. The quantity of cigarettes smoked ranged from 1 to 3 packs a day for 22 to 40 years and 10 cigars a day for 10 years.

Table 5. Polonium 210 in human organs, picocuries per gram of wet tissue of smokers and nonsmokers

Specimen	Number of specimens	Smoker 1	Number of specimens	Nonsmoker 1	Ratio of smoker to nonsmoker
Lung: Parenchyma Bronchi Nodes Liver Kidney Heart Psoas	8 7 7 6 6 6 3	0. 0079 . 0128 . 1070 . 0143 . 0088 . 0019 . 0006	1 4 4 3 3 3 2	0. 0025 . 0099 . 0380 . 0060 . 0057 . 0010	3. 16 1. 29 2. 82 2. 38 1. 54 1. 90 . 60

¹ Average values.

Our tests appeared to indicate that the level of polonium 210 is higher in four of the five organs tested from smokers than nonsmokers, except for the psoas muscle. Values for lung parenchyma agree well with the values obtained by Little and associates (12) for smokers and nonsmokers. We realize that in some instances the sample population is not large enough to yield significant results and can demonstrate only a trend.

Summary and Conclusions

The polonium 210 in tobacco may be implicated in the origin of lung cancer. Speculation on this possibility prompted a study at the Northeastern Radiological Health Laboratory, Public Health Service, to determine the levels of polonium 210 in several brands of cigarettes. The levels of lead 210 and radium 226 were also measured to determine whether the polonium 210 was in equilibrium with these precursors or was present independently. Samples of human organs were also analyzed for polonium 210 content to determine whether a difference could be detected in the concentration of this radionuclide between smokers and nonsmokers.

Polonium 210 and associated radionuclides lead 210 and bismuth 210 were found to be present in tobacco. Considerably smaller quantities of radium 226 were also found. Our tests indicated that lead 210 is deposited in the tobacco independently of radium 226 and its daughter

bismuth 210. For the various brands tested, the activities of polonium 210 in the total cigarette were of about the same level, with one exception: the cellulose plus charcoal brand was 30 percent higher than the average of all brands.

Because polonium 210 is volatile at the temperature of burning tobacco and subsequently is inhaled in the smoke, a smoking machine was devised for determining amounts inhaled from various brands. Values for polonium 210 in inhaled smoke ranged from 11 percent to 35.7 percent of that in the total cigarette. Average dosages to the lungs of a person smoking two packages of cigarettes per day were calculated to be far below the maximum permissible concentration for polonium 210 in air per person in the general population. The intake from a cellulose-filtered cigarette is 58.5 percent and from a filtered treated-tobacco cigarette, 45.1 percent of that from a nonfiltered cigarette.

To determine the feasibility of studying the distribution of polonium 210 in selected tissues and organs of the human body, random specimens of lung, liver, kidney, heart, and psoas muscle were obtained from smokers and non-smokers. Tests indicated higher levels of polonium 210 in the organs of smokers.

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Cutting the Death Rate From Cervical Cancer

Cervical cancer tests for the more than 8 million American women over 25 who are admitted to hospitals each year will be the first major goal of a multimillion dollar grant program. Within the next 5 years the Public Health Service expects to achieve this goal as the number of hospitals providing this service to all women patients increases each year during this period.

This campaign to eliminate deaths from cervical cancer is the first direct action program to result from recommendations of the President's Commission on Heart Disease, Cancer, and Stroke.

Hospitals providing care for the poor and medically indigent will be given first consideration in the awarding of grants.

A supplemental appropriation signed by the President provides \$5 million in grants to be awarded to hospitals during the first year of the program. This amount is expected to be increased by \$2.5 million the following 2 years.